PRELIMINARY PROSPECTUS

ADSORPTION AND DESORPTION STUDIES OF AMERICIUM AND PLUTONIUM IN WATER-SEDIMENT SYSTEMS AT ROCKY FLATS, COLORADO

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ADSORPTION AND DESORPTION STUDIES OF AMERICIUM AND PLUTONIUM IN SEDIMENT-WATER SYTEMS AT ROCKY FLATS, COLORADO**

INTRODUCTION

When radionuclides enter aquatic environments at Rocky Flats, they have several possible geochemical fates. They may remain in solution and be transported with the water mass or they may adsorb to organic or inorganic particulate matter and be transported with the suspended load or deposited in bottom sediments. During the three decades of plant operations, small amounts of plutonium have been released with process and sanitary waste discharges. Historically, all waste water has been treated in a sanitary or process treatment facility. After treatment, all water discharged from the plant flowed through a series of holding ponds, where settling of solids could occur prior to final release of the water to streams which flowed into municipal drinking water reservoirs. While deposition and permanent retention of transuranic elements on bottom sediments is the most beneficial scenario for these retention ponds, it cannot be assumed to occur under all potential environmental perturbations . A CSU study indicated a rapid transfer of plutonium from water to sediment, and core analyses revealed that the pond sediments were the primary site of deposition. However, a RF study on plutonium-bearing sediments from Pond-B1 demonstrated enhanced Pu-desorption at high pH presumably due to dispersion and repulsion of colloidal material. Quantification of such physico-chemical phenomena is important to insure containment of waste-bearing sediments during their current isolation and during future removal and decontamination operations.

** This is an outline prospectus for a research project that will be conducted during the summer of 1983 with the assistance of a summer intern student.

The focus of this proposed study is on adsorption/desorption processes involving sedimentary materials collected from the Rocky Flats plantsite. Adsorption/desorption from suspended particulates affects the partitioning between soluble and particulate phases and thus regulates the mobilty and ultimate fate of transuranic elements in the RFP environs. The experimental approach employed in this study will be to determine the distribution of americium and plutonium between the aqueous and particulate phases in laboratory sediment-water systems and then calculate K_d values* for different RF soils/sediments under varying conditions. This approach will be applied to uptake experiments on background soils/sediments (Lindsay Ranch/buffer zone) and desorption studies on contaminated sediments (Pond-B1). Such an applied approach will yield specific information on radionuclide removal processes which can used to assist in containment/decontamination operations and also provide predictive capabilities in the event of a catastrophic occurrence (i.e. large release of a strong acidic/basic discharge to retention ponds).

OBJECTIVES

The objective of this study is to obtain information that can be used to predict the fate of two transuranic elements, plutonium and americium in RFP aquatic environments. This objective will be pursued in a three stage program designed to be completed in a 3-4 month timeframe to maximally utilize the assistance of a summer intern student.

(1) Characterization of representative soils/sediments from Rocky Flats

--sediments from all retention ponds (A,B,C series & LR) will be analyzed for their organic matter contents(C & N,humic and fulvic acids), pH , Eh , zeta potential and cation exchange capacities and particle sizes.

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* The distribution coefficient is a ratio that compares the quantity of radionuclides in the particulate phase to the quantitiy in solution.

(2) Adsorption of Am & Pu onto uncontaminated RF soils/sediments

--a series of sequential extraction studies will be conducted to assess uptake under different conditions of temperature, pH, sediment concentration. Degree of reversibility , effects of bacterial mediated reactions and sorption kinetics will also be evaluated.

(3) Desorption of Am & Pu from contaminated RF soils/sediments

--a series of sequential extraction studies will be conducted to determine the extent and ease of desorption in the presence of different solutions (i.e. acid/base/EDTA/humic-fulvic acid, DDW, RFP-groundwater & surface water) over a range of environmental temperatures . Soils from the pad field and sediments from Pond B-1 (of known activities) will be used in these experiments.

SUMMARY-METHODOLOGY

The experimental procedures used in this research study will be adapted from published reports of other workers and modified as needed for the planned experiments on Rocky Flats soils and sediments. In particular, the methods developed by E.K. Duursma and his colleagues at the International Atomic Energy Agency (IAEA) Marine Laboratory will be implemented. All adsorption/desorption studies will utilize the "constant shaking technique" to determine distribution coefficients in laboratory sediment-water systems. Briefly the method consists of :

a. Adding the radionuclides to a known volume of filtered (0.45 & 0.22 um) water samples and adjusting the pH to the initial pH of the water sample with either dilute NaOH or HCl. Americium-241 and Plutonium-237 will be used in their soluble nitrate forms.

b. Adding dry sediments/soils from a stock sediment suspension to make a final predetermined sediment concentration, approximately 200 mg/l.

c. Shaking the sediment-water mixture in an agitating water bath held at a constant temperature.

d. Collecting samples at designated time intervals (6,12, 24,36,48,60,72....168) during the experiment,filtering (0.45 um) to separate particulate and dissolved radionuclides and measuring the concentration of radionuclides in the dissolved (dpm/ml) and particulate (dpm/gm) phases. The particulates on the filters will be weighed (g/ml) to correct for the units in determining the distribution values.

Using this technique large amounts of sediments can be exposed to the radionuclide solutions and the effects of sediment concentration can also be measured. For desorption studies the experiments will be performed in the same manner as the sorption experiments(using the various cocktails previously outlined). The degree of reversibility on all adsorption experiments will be verified . After a sorption experiment is completed (7 days), the sediment sample will be centrifuged and the radioactive solution above the sediment will be decanted and replaced with an equal amount of unspiked, distilled,deionized water. The reversibility study will then proceed to term. Effects of microbially-mediated uptake/adsorption will be evaluated by using aliquots of sterilized sediments. Irradiation of the sediments with 10^6 Rads from a ^{60}Co irradiator should minimize bacterial growth over the 7 day extraction term of the experiments.

COUNTING TECHNIQUES

Experimental samples will be counted using a Ge(Li) system to measure the low energy X- and gamma rays of ^{237}Pu and ^{241}Am. In some cases it would also be advantageous to use a NaI well crystal for counting; because of the differences in energies of the ^{237}Pu (97-101 Kev) and ^{241}Am (60 Kev), the NaI could be used to resolve the energies of these two isotopes in dual-labeled experiments.

REQUIREMENTS

--minimal lab space for duration of experimental work(i.e. bench space area such as in room 111/building 123)

-- pH meter (acquired)

-- zeta potential meter (acquired)

-- radiotracers (can get Am-241 from standards lab)

-- glassware (partially acquired)

-- pipettes,lab supplies (acquired)

-- agitating water bath

Availability of the lab space needs to be worked out; total acquisition costs for the project would be approximately $2500. (agitating water bath-$1200; radiotracer(Pu-237)$1000; glassware-$300). An attempt is being made to acquire an agitating water bath which would reduce project costs by one-third.